Neutron Scattering Group(Annual Report)

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Neutron Scattering Group

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Research Activities

In this period of 1989 we have completed the construction of the new neutron scattering spectrometer which is installed in the 6G beam hole of the JRR3 reactor at the Tokai establishment of JAERI. The spectrometer was designed such that the built-in polarizer can be switched in any time necessary to change from unpolarized to polarized neutron scattering mode. The spectrometer has been constructed by the NKK Co.Ltd. jointly worked with the

Rigaku and Kohzu Industry Co.. The polarizing instrumentation has been constructed by Tokin Industry Ltd.. Since the polarizer itself is now developing under the collaboration project with Institute Laue Langevin (Dr.A.Magerl), it needs some years until we are able to work with the perfect operation of polarized neutron beam. The system is now under the final test and it is complicated, since the main movable component is cussioned by air blow onto the marble floor which requires a precise control of the delicate as well as sequencial coordination of every parts. After finishing this test we plan to perform a bench mark experiment and to construct the computer network connected to both experimental base at Tokai and the university office in Sendai.

The main activity outcomes are summarized here as usual by listing the principal investigators for each subject. Several small subjects are not listed in this report which are more or less pursued on the individual basis.

(I) **High Tc Superconductivity** (Priority Field in Scientific Research for the Grant in Aid)

a) Two dimensional antiferromagnetic spin correlations in undoped and Ce doped Nd_2CuO_4 and Pr_2CuO_4

(M.Matsuda,T.R.Thurston,K.Takada,K.Yamada,K.Kakurai and Y.Endoh)

Two dimensional (2D) antiferromagnetic spin correlations in undoped and Ce doped Nd_2CuO_4 and Pr_2CuO_4 have been studied by neutron scattering measurements. For all the compounds we have firstly observed well defined 2D magnetic scattering above T_N corresponding to the strong inplane antiferromagnetic correlation between Cu spins. The correlation lengths and their temperature variation for undoped compounds are almost identical to those of La_2CuO_4.

Even in the Ce doped system the correlation length exhibits a remarkable temperature dependence in contrast to the case of Sr doped La_2CuO_4 where the correlation length is almost temperature independent probably due to the frustrated interaction between Cu spins induced by the hole spins. These experimental results suggest different carrier site or different degree of the carrier localization between two types of cuprates, hole and electron carrier type. Studies in the superconducting state are now developing using the heat treated crystals.

This subject is collaborated with several groups: Brookhaven National Laboratory (Dr. G.Shirane, Dr. P.Gehring), Massachusetts Institute of Technology (Prof. M.Kastner and Prof. R.J.Birgeneau), NTT Optoelectronics Laboratories (Dr. Y.Hidaka) and Yamanashi University (Prof.H.Kojima and Dr.I.Tanaka).

b) Magnetic structures in Nd_2CuO_4 and Pr_2CuO_4 with T'-type tetragonal structure

(M.Matsuda,T.R.Thurston,K.Takada,K.Yamada,K.Kakurai and Y.Endoh)

The three dimensional (3D) antiferromagnetic structures have been studied in Nd_2CuO_4 and Pr_2CuO_4 by neutron scattering measurements. In Nd_2CuO_4 successive magnetic phase transitions due to the inplane spin reorientations of Cu^{2+} ions have been observed. This phase transitions are similar to those obsreved in La_2CoO_4 where the spin reorientations are known to be triggered by the structural phase transition from the orthorhombic to the so called low temperature tetragonal phase. However in Nd_2CuO_4 despite of the detailed examinations by not only neutron but also X-ray measurements, no structural change or deformation from the T'-type tetragonal symmetry has been detected through the transition temperatures. In Pr_2CuO_4, such successive phase transitions have only been observed under pressure.

In both cuprates, the rare earth spins participate in the 3D antiferromagnetic order. We have determined the magnetic moments of Cu and the rare earth ions in the 3D ordered phase. Due to the interference effect on the magnetic structure factor small magnetic moment of about 0.03 μ_B/Pr is detectable in Pr_2CuO_4.

By doping Ce ions the Neel temperatures decrease substantially but the 3D order has been clearly observed with doping up to x=0.075 in both $Pr_{2-x}Ce_xCuO_4$ and $Nd_{2-x}Ce_xCuO_4$. The heat treatment effects on the stability of the 3D magnetic order have also been studying.

This subject is collaborated with several groups: Brookhaven National Laboratory (Dr. G.Shirane, Dr. P.M.Gehring), Massachusetts Institute of Technology (Prof. R.J.Birgeneau), NTT Optoelectronics Laboratories (Dr. Y.Hidaka) and Institute for Materials Research, Tohoku University (Y.Watanabe).

c) Magnetization process in superconducting $La_{2-x}Sr_xCuO_4$
(T.Watanabe,S.Itoh,K.Kuroda and Y.Endoh)

Time-of-flight polarized neutron transmission measurements have been performed from single crystals of oxygen superconductor $La_{1.85}Sr_{0.15}CuO_4$ and V_3Si, one of the typical type-II superconductor. Qualitative analyses of the neutron wave length dependence of the depolarization provide valuable information such as lower critical field H_{c1} and its anisotropy and the spatial distribution of the trapped magnetic flux in the superconducting phase. The anisotropic H_{c1} has been compared with that obtained by the magnetization measurement and the origin of the small difference in the H_{c1} obtained by the different methods will be studied to clarify the flux pinning state of the oxygen superconductor. Collaborated Groups are Massachusetts Institute of Technology (Prof.R.J.Birgeneau), NTT Optoelectronics Laboratories (Dr.Y.Hidaka), Yamanashi University (Prof.H.Kojima, Dr. I.Tanaka), and Institute for Material Research (Prof.N.Toyota)

d) Growth of large single crystals of $La_2NiO_{4+\delta}$ and $La_2CoO_{4+\delta}$ and control of the oxygen stoichiometry

(K.Yamada,K.Nakajima,Y.Noda,S.Onodera and Y.Endoh)

Single crystals of $La_2NiO_{4+\delta}$ with the dimensions of about 8mm^{f}x 60mm have been prepared by the floating zone method in air. The as-grown crystal has tetragonal symmetry at room temperature and has very low Neel temperature T_N. For the convenience of the spin wave measurements , the crystals are annealed in CO_2+CO (0.1%) atmosphere to raise the T_N to around 200K. It is found that the T_N is very sensitive to the oxygen stoichiometry which is controlled reversibly by the oxygen partial pressure during the annealing and the annealing time.

In the case of $La_2CoO_{4+\delta}$ although the similar size of the single crystal as $La_2NiO_{4+\delta}$ can be prepared, the crystal collapses into small pieces within a week probably due to the change of the oxygen stoichiometry. We are now developing the automatic oxygen pressure controller coupled with the zirconium oxygen sensor to find out the best annealing condition for $La_2CoO_{4+\delta}$.

This subject is collaborated with Dr S. Hosoya in Institute for Material Research.

(II) Dense Kondo systems and anomalous rare earth compounds

a) Magnetic phase diagram of $PrCo_2Si_2$ and $TbNi_2Si_2$

(M. Osada, K. Takada, H. Kawai, K. Kakurai, K. Yamada and Y. Endoh)

In these antiferromagnetic(AF) rare earth compounds of $ThCr_2Si_2$ type with body centred tetragonal structure the moments on rare earth ions have strong Ising anisotropy. When the field is applied along the easy c-axis,the magnetization reveals discrete steps before reaching the saturation moment, i.e. they show a characteristic metamagnetic behaviour. Neutron scattering studies have demonstrated the existence of complicated magnetic structures of large unit cells with temperature and field.

$PrCo_2Si_2$: In zero magnetic field three magnetic phases below T_N were known. Since the k-vector of the antiferromagnetic order is parallel to the easy axis direction, we had to use the horizontal field configuration with the limited maximum field of 16.3kOe, just enough to reach the first magnetization step at temperatures $T \geq 8K$. We mapped the temperature and field phase diagram and found that the phase boundary temperatures between the different AF ordered phases decrease with increasing field. The neutron diffraction experiments are now extended to the high magnetic field by using the pulsed field technique synchronized with the pulsed neutrons.

$TbNi_2Si_2$: Here the k-vector of the AF ordering is perpendicular to the easy axis direction. Hence the vertical field up to 48kOe could be applied. At low temperature (T=2K) the main six spin structures associated to the different magnetization steps could be determined. These structures can be understood by assuming a strong 1-D ferromagnetic(FM) exchange along the c-axis, an intraplane nearest neighbour AF and next nearest neighbour FM exchange and a weak interplane AF exchange. Indeed strong one-dimensional spin correlations along the c-axis above T_N has been observed by quasielastic neutron scattering. Whether the numerous incommesurate structures observed at elavated temperatures can be interpreted within this competing exchange model is the subject of the future study.

This work is performed in collaboration with Yamaguchi University (Dr. T. Shigeoka), Hiroshima University (Prof. H. Fujii), Kobe University (Prof. H.Motokawa and Dr. Nojiri) and Brookhaven National Laboratory (Dr. L. Rebelsky and Prof. G. Shirane).

b) Neutron scattering studies of ferromagnetic Kondo system $CeSi_x$
(M. Kohgi, K. Ohoyama)

The intermetallic compound $CeSi_x$ shows the typical heavy electron anomalies for $1.83 < x \leq 2$. It is ferromagnetic for $1.6 < x \leq 1.83$ with the same crystal structure. The ferromagnetic instability at around x=1.83 is considered to originate from the competition between the Kondo effect and the inter-atomic exchange interactions. We are studying this system by means of neutron scattering in order to make clear the origin of the systematic change of the electronic state with changing x. The experiments were carried out on the chopper spectrometer INC at KENS, KEK. We found that the linewidths of the crystal field excitations in $CeSi_x$ are anomalously large and increase with increasing x and that their size are comparable to the high energy scale Kondo temperature $T_K^{(6)}$, which is defined as the Kondo temperature for the six-fold ground multiplet of Ce^{3+} ion, of this system. The result gives another support to the idea that the x-dependence of the hybridization of 4f electrons with conduction electrons is the main origin of the phase change from the ferromagnetic to nonmagnetic Kondo state.

This subject is studied in collaboration with T. Satoh, Tohoku University, M. Arai, National Laboratory for High Energy Physics.

c) Neutron scattering study of Yb monopnictides
(K. Ohoyama, M. Kohgi and T.Osakabe)

The NaCl type compounds Ytterbium monopnictides YbX (X=N, P, As) order antiferromag-netically below 1 K with rather small saturation sublattice moment of less than about 0.8 μ_B, which are fairly small compared with the ionic value of about 2 μ_B for the ground state doublet. They show also a broad hump in the specific heat around 5 K which is typical of Kondo system with T_K of about 5 K. In order to make clear the origin of these anomalous phenomena, we are studying these compounds by means of neutron scattering mainly at the chopper spectrometer INC at KENS, KEK. We observed the clear inelastic peaks corresponding to the transition between the ground and first excited states at about 34, 23 and 19 meV for x=N, P, and As, respectively. For the position of the second excited state, it is at 42 meV for YbAs case, however further investigation is neccessary for the other cases because of the difficulty to distinguish the magnetic scattering from the phonon background. Such work as well as the investigation of the quasi-elastic scattering are now under way.

This subject is studied in collaboration with T. Suzuki and A. Oyamada, Tohoku University, M. Arai, National Laboratory for High Energy Physics.

(III) 3d-Itinerant magnetism

a) Magnetic properties in CoS_2
(H.Hiraka, K.Yamada and Y.Endoh)

Neutron critical diffuse scattering and magnetization measurements have been performed from single crystal of CoS_2 prepared by the chemical tranport technique. We have determined the effective exchange constant between Co spins from the analysis of the temperature dependence of the spontaneous magnetization. The magnetic correlation length ξ obtained from the line width of the diffuse scattering diverges at T_C. The correlation length is much larger than those of another isotropic ferromagnets such as EuS, EuO and Fe when compared at the same reduced temperatures. Detailed data analysis for the discussion of the critical index ν is now in progress.

(IV) Spin glass

a) Re-entrant spin glass properties in FeAl and AuFe alloys
(J.Suzuki,T.Watanabe,S.Itoh and Y.Endoh)

The history dependent phenomenon is a major characteristics of the spin glass. Furthermore the magnetic response to the external conditions is dependent on their frequency; such as the cusp in thermal evolution of the AC susceptibility shifts tremendously with the frequency as well as the external field strength. These concomitant properties make the definitive experiments as well as the unique interpretations very difficult. Thus the previous experiments have been reexamined in the different view by using the different neutron techniques such as pulsed neutron small angle scattering, depolarization measurements and so forth for the classical metallic spin glass of FeAl and AuFe. Already the important informations indicating the exsistence of a kind of mixed phase as predicted by the mean field theory have been observed, which is now analyzed. The similar novel neutron techniques have been applied to the magnetically heterogeneous materials such as the ferrofluid which is the colloid of the forromagnetic ultrafine particles suspended in the organo solution. The experiments indicated that we can consider the thermodynamical behavior on the basis of the fundamental physics.

This work is collaborated with ISSP (Prof.H.Yoshizawa and Dr. S.Mitsuda) and CEN Sacray (Dr.I.Mirebeau)

(V) Low dimensional magnetism

a) Quantum effect in quasi one-dimensional antiferromagnets
(K. Kakurai, K. Nakajima and Y. Endoh)

We have been studying the spin dynamics of the S=1, quasi one-dimensional antiferromagnetic systems $CsNiCl_3$, $CsNiBr_3$ and $RbNiCl_3$. In all three systems the spin wave(SW) dispersions even in the 3-D ordered phase cannot be consistently described by the classical SW theory based upon the commenly accepted Hamiltonian. The discrepancy can be solved if the Haldane gap is introduced for the individual chains whose character then is kept even in the ordered state. The quantitative agreement of the required gap values in all three systems with the Haldane prediction strongly suggests the importance of the quantum effect in these 3-D ordered quasi one-dimensional antiferromagnets. The recent high field measurement using polarized neutron scattering supports the above interpretation.

We continuously study on the spin dynamics of the S=1/2, one dimensional spin dynamics which is the extension of the previous experimental studies from CPC. Since the enrgy scale of CPC is not best suited for the further study of spin dynamics, a new material of the archetype of the 1D antiferromagnets have been looked for. Quite recently a new candidate is found and now the detailed experimants are going on by using this new material.

The new nonlinear spin excitation modes under the transverse external field have been observed from the TMMC.

These works are carried out in collaboration with University of Mainz (Prof. M. Steiner), Riso National Laboratory (Dr. J.K. Kjems) , Institut Laue-Langevin (Dr. R. Pynn), Tokyo Institute of Technology (Dr. H. Tanaka and Prof. K. Iio), CEN Grenble (Dr. J.P.Boucher and L.P. Regnault) and Institute for Material Science (Dr.S.Hosoya).

(VI) Polarized neutron works

a) Small angle polarized neuton scattering and Pulsed field neutron diffraction
(S.Itoh, T.Watanabe, H.Kawai, K.Yamada and Y.Endoh)

As the modernization of the KENS facility as well as the preliminary studies for the next generation pulsed polarized neutron project, we are developing the new neutron scattering method which has never been exsisted although a lot of discussions were made. These are the polarized neutron small angle scattering camera with which only the magnetic scattering component is separated out from the total small angle scattering. The transverse and pararell responses to the magnetization are separated by the polarization anlysis. For this purpose we attach the built in analyser component in front of the detector.

As for the pulsed field measurement we set the repetitive pulsed magnet with the frequency of 1/2 Hz, and the maximum field of about 20Tesla. The field is produced by the discharge of the large bank of the condensor every 2 second coincided with the neutron pulses and within 2 seconds it is energized. The coil is designed such that the coil is continuously used without any damage for more than about 100 thousand cycles. The work is carried out by the collaboration with Kobe University (Prof. H.Motokawa and Dr. Nojiri), KEK (Dr. M.Arai and M.Furusaka)

b) Application of the crosscorrelation technique for the pulsed polarized white neutron beam (H. Fujimoto and M. Kohgi)

We have finished the development of the crosscorrelation technique for the pulsed polarized white neutron beam on the PEN spectrometer at KENS, KEK. The method relies on pseudorandom modulation of the polarization of the incident neutron beam. Subsequent crosscorrelation of the detected count rate with the pseudorandom modulating sequence yields a two dimensional spectrum of the difference between the spin flip and non flip cross sections of the sample at a wide range combination of incident and final energies. To realize this method , we constructed a polarization modurating device system and a two dimensional time analyzer system. In order to test the performance of the present system, we measured spin wave scattering from the ferromagnetic Heusler alloy Cu_2MnAl. We could obtaine the TOF spectra of the spin wave scattering in a wide range of the scattering plane around 111 reciprocal lattice point. The observed spectra are in good agreement with the simulation calculation which takes account of neutron pulse shape, time uncertainty at the spin flipper and the angular diversion. The results of the present experiment show that the combination of the pulsed polarized white neutron beam and the crosscorrelation method with polarization modulation is a quite promising way for the study of magnetic excitations or fluctuations in various magnetic materials.

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Doctor Thesis

D1) Polarized neutron study of heterogeneous magnetic materials
(Shinichi Itoh) (March 1990)

Master Thesis

M1) Magnetic phase transitions in Ising antiferromagnets RM_2Si_2
(Masashi Osada) (March 1990)

M2) Construction of polarized neutron spectrometer
(Kenji Nakajima) (March 1990)

M3) Polarized neutron study of mixing states of high T_c superconductors
(Tsukasa Watanabe) (March 1990)